Connecting the dots: Reinventing optics for nanoscale dimensions

Naomi J. Halas[1]

Department of Electrical and Computer Engineering, Laboratory for Nanophotonics, Rice University, 6100 Main Street, Houston, TX 77005-1892

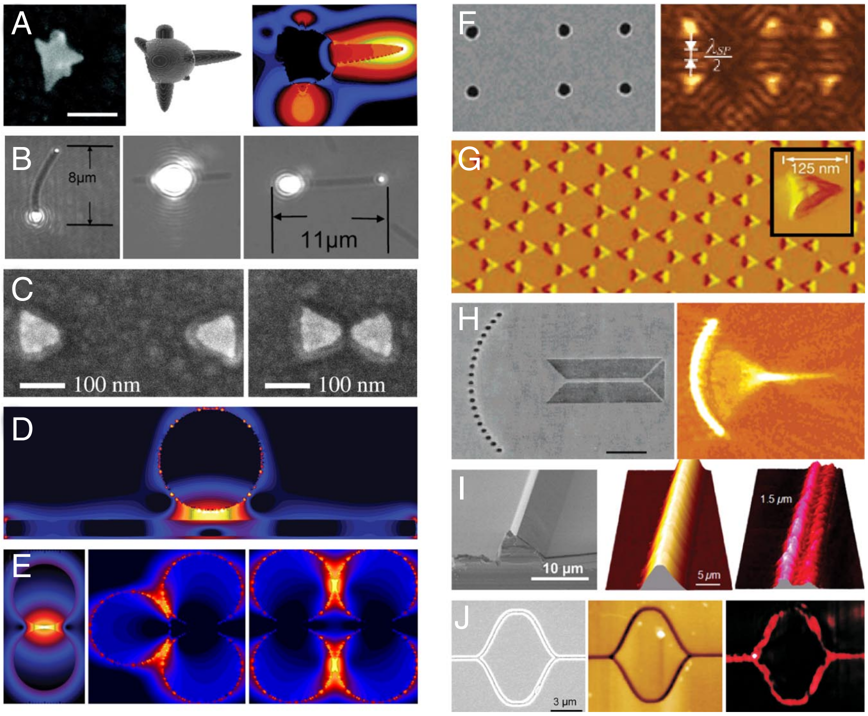

Fig. 1. A visual survey of nanoscale optical components. (*A*) Gold nanostar SEM image, simulation geometry, and electromagnetic simulation of their optical response using the finite difference time domain (FDTD) method. [Reproduced with permission from ref. 6 (Copyright 2007, American Chemical Society)]. (*B*) Silver nanowires as plasmon waveguides. [Reproduced with permission from ref. 7 (Copyright 2006, American Chemical Society)]. (*C*) Gold bowtie nanoantennas. [Reproduced with permission from ref. 11 (Copyright 2004, American Chemical Society)]. (*D*) FDTD simulation of a gold sphere over a thin gold film. (*E*) Nearly-touching and touching gold nanorod pairs and a gold nanoparticle pair. [Reproduced with permission from ref. 19 (Copyright 2006, American Chemical Society)]. (*F*) SEM and near-field scanning optical microscopy (NSOM) images of nanoholes in a gold film. [Reproduced with permission from ref. 14 (Copyright 2006, American Chemical Society)]. (*G*) Atomic force microscope (AFM) image of a nanoparticle array fabricated by using nanosphere lithography. [Reprinted, with permission, from page 273 of the *Annual Review of Physical Chemistry*, Volume 58, Copyright 2007 by Annual Reviews (www.annualreviews.org) (15)]. (*H*) SEM and NSOM images of an Au/Cr plasmon focusing array. [Reproduced with permission from ref. 16 (Copyright 2005, American Chemical Society)]. (*I*) SEM, AFM, and NSOM images of a subwavelength metal wedge waveguide. [Reproduced with permission from ref. 17 (Copyright 2008, Optical Society of America)]. (*J*) SEM, AFM, and NSOM images of a plasmonic Mach–Zehnder interferometer made by using V-groove waveguides. [Reprinted by permission from Macmillan Publishers Ltd: *Nature* (18), copyright (2006)].

While optics is one of our oldest scientific tools, enabling some of the earliest advances in astronomy and biology, it is also currently one of the most dynamic and exciting areas of applied science. Developments of the past two decades in nanoscale device fabrication, nanomaterials synthesis and patterning, and advanced computational modeling capabilities have converged to fuel a revolution in optical science, leading to an entirely new tool set for optics at nanometer length scales. The work described in a recent issue of PNAS (1) illustrates the innovative use of nanoparticles as sensitive optical tools that provide a new way to measure the properties of light at nanometer-scale dimensions.

To the casual user of optics, the idea of optical tools at nanoscale dimensions seems oxymoronic. After all, aren't all optical imaging systems restricted by the diffraction limit of light, the seemingly universal restriction that limits our ability to focus light and therefore resolve images smaller than an optical wavelength? Although the diffraction limit clearly holds for classical imaging, in the past two decades a wealth of new strategies that allow us to circumvent the diffraction limit and manipulate light at dimensions far below that of an optical wavelength have been developed. Many of these approaches exploit the unique properties of metals to support electromagnetic waves at their surfaces, through the oscillation of their conduction electrons known as surface plasmons. With this approach, an interesting analogy and scaling principle emerges. Just as radio-frequency antennas provide sources of electromagnetic waves and are much smaller than the wavelengths of radiation they emit, the same principle holds for visible light and nanoscale metallic structures, which serve as tiny nanoscale antennas for the much larger wavelengths of emitted, or scattered, light. Radio-frequency antennas can both transmit and receive signals, and analogously, optical nanoantennas (2, 3) can also serve as transmitters and receivers, collecting, focusing, guiding, and manipulating light in a variety of novel ways. This general principle forms the basis for many current advances in nanoscale optics and optical design, leading to new types of imaging and fabrication tools. This approach has also spawned the field of metamaterials, which incorporates nanoantenna-like structures into materials to impart new optical properties not found in the materials nature provides (4, 5).

Just as radio-frequency antennas exist in a wide variety of shapes, sizes, and orientations for a multitude of uses, at the nanoscale the geometry and orientation of metallic nanoantennas control their properties and the types of applications for which they are most suited. Virtually any type of metallic nanostructure can serve as a nanoantenna and interact with light: the size, geometry, and orientation of the structure itself controls the light–nanoantenna interac-

---

Author contributions: N.J.H. wrote the paper.

The author declares no conflict of interest.

See companion article on page 2495 in issue 8 of volume 106.

[1] E-mail: halas@rice.edu.

www.pnas.org/cgi/doi/10.1073/pnas.0900796106

tion. This simple property has led to an extraordinary proliferation of various geometries of metallic nanoparticles and nanostructures that has fascinated chemists, physicists, and engineers alike (Fig. 1). Historically, some of the beautiful colors of stained-glass windows originating in antiquity result from embedding metallic nanoparticles of gold and silver into glass. These origins have inspired modern-day chemists to develop metallic nanoparticles of a vast variety of shapes and sizes that preferentially absorb or scatter light at wavelengths determined by shape and that are fabricated by chemical means (Fig. 1 *A* and *B*) (6, 7). This approach has even reached the realm of biomedical technology, where nanoparticles designed to absorb or scatter light at near infrared wavelengths, transmissive in the human body, developed as in vivo probes for diagnostics and therapeutics (8). Extraordinary and highly complex nanoparticles, such as "nanostars" (6) or long, single-crystalline nanowires (7), can be fabricated by chemical methods. Both of these structures couple strongly to light. In the nanostar, the central core of the nanoparticle acts as an antenna, transmitting the resonant frequencies determined by the lengths and positions of the asperities of the structure. Nanowires act as highly polarization-sensitive transmitters and nanoscale optical waveguides. Another complementary fabrication approach has harnessed the powerful clean-room patterning and nanofabrication tools of the semiconductor industry to fabricate efficient bowtie nanoantennas (9) onto planar substrates with highly-precise geometries (Fig. 1*C*). These structures are best known for providing a method for focusing light into the nanoscale junction between conductors, resulting in extremely high optical intensities in this junction region. By positioning molecules in this junction region, molecular spectroscopy of single molecules is achievable (9–11). Hybrid fabrication methods that combine both chemical and physical processes to build complex nanostructures not achievable by wet or dry methods alone have been pioneered to fabricate new structures and to position structures precisely in complex patterns, over large areas inexpensively, and in complex periodic arrays (Fig. 1 *D–G*) (12–15).

Virtually any type of metallic nanostructure can serve as a nanoantenna.

Nanoantennas positioned in specific geometries can be used for directional light-guiding, in direct analogy with radio-frequency antenna arrays. Nanoantenna arrays have been shown to couple light directly into nanoscale metallic waveguides with cross-sections far smaller than conventional optical fibers (16). Waveguide-based devices at these dimensions have been demonstrated (17). New and innovative waveguide geometries being developed address issues such as propagation losses and provide useful geometries that may lead, for example, to active light-based logic devices (18).

In the article by Cubukcu et al. (1), the nanoantenna paradigm is applied in 2 different ways, in types of nanostructures that function in a unique receiver–transmitter relationship. The "receivers" are gold nanodisks that couple to input light at their characteristic resonant frequency. The input light excites resonant oscillations in these structures, setting up well-defined electromagnetic modes with a complex field pattern. Adjacent to the nanodisks are single-walled carbon nanotubes that have been precisely aligned through directional growth on the substrate, before the patterning of the nanodisks. The nanotubes themselves are also nanoantennas, acting as "transmitters" in the experiment. When the nanodisk receivers are excited, their local optical field couples to the adjacent nanotubes. This coupling is highly directional, with the nanotubes responding to the local field of the nanodisks when the field is along the direction of the nanotube axis, thus functioning as polarization-dependent, near-field detectors. The local fields of the nanodisks excite the Raman vibrational modes of the carbon nanotubes, providing a unique characteristic signature to the optical signal that the carbon nanotube antennas transmit to the far field, which is ultimately detected as the output signal.

These beautifully-executed arrays of transmitter–receiver nanoantenna pairs provide a new direction in nanoantenna development and in optics at nanoscale dimensions. Combining carbon nanotubes, essentially molecular nanowire antennas, with metallic nanoantennas, links 2 material systems that provide complementary functions and enable the transmitter–receiver pair concept to be realized in a practical manner. The combination of materials and structures also eliminates unwanted antenna coupling or cross-talk, because the information flow is directional from nanodisk to nanotube because of the difference in optical cross-sections of the 2 disparate types of structures. Chemical functionalization of the nanotubes or geometrical variation of the nanodisks could provide a network of receiver and reporter nodes that would be both uniquely optically addressable and uniquely identifiable by the spectrum of the output signal. This type of structure could ultimately provide optical readout for on-chip nanophotonic logic or routing devices. It seems inevitable that this bold new advance in nanoantenna design and function will lead to new and exciting developments in the field of nanoscale optics.

1. Cubukcu E, et al. (2009) Aligned carbon nanotubes as polarization-sensitive, molecular near-field detectors. *Proc Natl Acad Sci USA* 106:2495–2499.
2. Alu A, Engheta N (2008) Tuning the scattering response of optical nanoantennas with nanocircuit loads. *Nat Photonics* 2:307–309.
3. Alu A, Engheta N (2008) Input impedance, nanocircuit loading, and radiation tuning of optical nanoantennas. *Phys Rev Lett* 101:043901.
4. Veselago VG (1968) The electrodynamics of substances with simultaneously negative values of epsilon and mu. *Sov Phys Uspekhi* 10:509–514.
5. Linden S, et al. (2004) Magnetic response of metamaterials at 100 Terahertz. *Science* 306:1351–1353.
6. Hao F, Nehl CL, Hafner JH, Nordlander P (2007) Plasmon resonances of a gold nanostar. *Nano Lett* 7:729–732.
7. Sanders AW, et al. (2006) Observation of plasmon propagation, redirection, and fan-out in silver nanowires. *Nano Lett* 6:1822–1826.
8. Lal S, Clare SE, Halas NJ (2008) Nanoscience-enabled cancer therapy: Impending clinical impact. *Acc Chem Res* 41:1842–1851.
9. Schuck PJ, Fromm DP, Sundaramurthy A, Kino GS, Moerner WE (2005) Improving the mismatch between light and nanoscale objects with gold bowtie nanoantennas. *Phys Rev Lett* 94:017402.
10. Moskovits M (1985) Surface-enhanced spectroscopy. *Rev Mod Phys* 57:783.
11. Fromm DP, Sundaramurthy A, Schuck PJ, Kino G, Moerner WE (2004) Gap-dependent optical coupling of single "bowtie" nanoantennas resonant in the visible. *Nano Lett* 4:957–961.
12. Nordlander P, Le F (2006) Plasmonic structure and electromagnetic field enhancements in the metallic nanoparticle-film system. *Appl Phys B* 84:35–41.
13. Willingham B, Brandl DW, Nordlander P (2008) Plasmon hybridization in nanorod dimers. *Appl Phys B* 93:209–216.
14. Gao H, Henzie J, Odom TW (2006) Direct evidence for surface plasmon-mediated enhanced light transmission through metallic nanohole arrays. *Nano Lett* 6:2104–2108.
15. Willets KA, Duyne RPV (2007) Localized surface plasmon resonance spectroscopy and sensing. *Annu Rev Phys Chem* 58:267–297.
16. Yin L, et al. (2005) Subwavelength focusing and guiding of surface plasmons. *Nano Lett* 5:1399–1402.
17. Boltasseva A, et al. (2008) Triangular metal wedges for subwavelength plasmon-polariton guiding at telecom wavelengths. *Optics Express* 16:5252–5260.
18. Bozhevolnyi SI, Volkov VS, Devaux ES, Laluet J-Y, Ebbesen TW (2006) Channel plasmon subwavelength waveguide components including interferometers and ring resonators. *Nature* 440:508–511.
19. Brandl DW, Mirin NA, Nordlander P (2006) Plasmon modes of nanosphere trimers and quadrumers. *J Phys Chem B* 110:12302–12310.